LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

UCRL-JRNL-207122

Clocking Femtosecond X-Rays

A. L. Cavalieri, D. M. Fritz, S. H. Lee, P. H. Bucksbaum, D. A. Reis, D. M. Mills, R. Pahl, J. Rudati, P. H. Fuoss, G. B. Stephenson, D. P. Lowney, A. G. MacPhee, D. Weinstein, R. W. Falcone, J. Als-Nielsen, C. Blome, R. Ischebeck, H. Schlarb, T. Tschentscher, J. Schneider, K. Sokolowski-Tinten, H. N. Chapman, R. W. Lee, T. N. Hansen, O. Synnergren, J. Larsson, S. Techert, J. Sheppard, J. S. Wark, M. Bergh, C. Calleman, G. Huldt, D. van der Spoel, N. Timneanu, J. Hajdu, E. Bong, P. Emma, P. Krejcik, J. Arthur, S. Brennan, K. J. Gaffney, A. M. Lindenberg, J. B. Hastings

October 8, 2004

Physical Review Letters

Clocking Femtosecond X-Rays

A. L. Cavalieri,[1] D. M. Fritz,[1] S. H. Lee,[1] P. H. Bucksbaum,[1] D. A. Reis,[1] D. M. Mills,[2] R. Pahl,[2] J. Rudati,[3] P. H. Fuoss,[3] G. B. Stephenson,[3] D. P. Lowney,[4] A. G. MacPhee,[4] D. Weinstein,[4] R. W. Falcone,[4] R. Pahl,[5] J. Als-Nielsen,[6] C. Blome,[7] R. Ischebeck,[7] H. Schlarb,[7] Th. Tschentscher,[7] J. Schneider,[7] K. Sokolowski-Tinten,[8] H. N. Chapman,[9] R. W. Lee,[9] T. N. Hansen,[10] O. Synnergren,[10] J. Larsson,[10] S. Techert,[11] J. Sheppard,[12] J. S. Wark,[12] M. Bergh,[13] C. Calleman,[13] G. Huldt,[13] D. van der Spoel,[13] N. Timneanu,[13] J. Hajdu,[13] E. Bong,[14] P. Emma,[14] P. Krejcik,[14] J. Arthur,[15] S. Brennan,[15] K. J. Gaffney,[15] A. M. Lindenberg,[15] & J. B. Hastings[15]

[1] *FOCUS Center, Departments of Physics and Applied Physics Program, University of Michigan, Ann Arbor, MI 48109, USA*

[2] *Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, USA*

3 *Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, USA*

[4] *Department of Physics, University of California, Berkeley, CA 94720, USA*

[5] *Consortium for Advanced Radiation Sources, University of Chicago, Chicago, IL 60637, USA*

[6] *Niels Bohr Institute, Copenhagen University, 2100 Copenhagen Ø, Denmark*

[7] *Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85, 22607 Hamburg, Germany*

[8] *Institut für Laser- und Plasmaphysik, Universität-GHS-Essen, D-45117 Essen, Germany*

[9] *Physics Department, Lawrence Livermore National Laboratory, Livermore, CA 94550, USA*

[10] *Department of Physics, Lund Institute of Technology, P.O. Box 118, S-22100, Lund, Sweden*

[11] *Max Plank Institute for Biophysical Chemistry, Am Faßberg 11, 37077 Göttingen, Germany*

[12] *Atomic and Laser Physics Department, Oxford University, Oxford OX1 2JD, UK*

[13] *Department of Biochemistry, Uppsala University, Biomedical Center, Box 576, S-751 23 Uppsala, Sweden*

[14] *Stanford Linear Accelerator Center, Menlo Park, CA 94025, USA*

[15] *Stanford Synchrotron Radiation Laboratory/SLAC, Stanford University, Menlo Park, CA 94025, USA*

The Sub-Picosecond Pulse Source (SPPS) at the Stanford Linear Accelerator Center (SLAC) produces the brightest ultrafast x-ray pulses in the world, and is the first to employ compressed femtosecond electron bunches for the x-ray source[1,2]. Both SPPS and future X-ray Free Electron Lasers (XFEL's) will use precise measurements of individual electron bunches to time the arrival of x-ray pulses for time-resolved experiments. At SPPS we use electro-optic sampling (EOS) to perform these measurements. Here we present the first results using this method. An ultrafast laser pulse (135 fs) passes through an electro-optic crystal adjacent to the electron beam. The refractive index of the crystal is distorted by the strong electromagnetic fields of the ultra-relativistic electrons, and this transient birefringence is imprinted on the laser polarization[3]. A polarizer decodes this signal, producing a time-dependent image of the compressed electron bunch. Our measurements yield the relative timing between an ultrafast optical laser and

an ultrafast x-ray pulse to within 60 fs, making it possible to use the SPPS to observe atomic-scale ultrafast dynamics initiated by laser-matter interaction.

Ultrafast x-ray pulses are providing our first view of atomic motion in both space and time. New sources based on high harmonic generation[4,5], laser-produced plasmas[6], or modulated electron accelerators[7] produce x-ray pulses with durations of less than a few hundred femtoseconds. This is the time-scale of vibrations in solids and molecules, and the making and breaking of chemical bonds. While these sources provide the time resolution to study these dynamics, experiments are often hindered by their relatively low brightness. XFEL's will be more than twenty orders of magnitude brighter than laser plasma sources with the potential for time-resolution at or below one femtosecond, short and bright enough to image chemistry in real time on the atomic scale. SPPS is intermediate in brightness between existing ultrafast x-ray sources and future XFEL's, and can be used to develop scientific techniques required for the success of future accelerator-based sources.

Sub-picosecond time-dependent phenomena are typically studied with pump-probe techniques in which the dynamics are initiated with an ultrafast laser or laser-driven source and then probed after a time delay. Generally, these experiments are self-synchronized because the pump and probe have a common laser source; therefore, precise time delays can be produced with different optical path lengths. The time resolution is then limited by the overlap of the pump and probe pulses, and can be as short as a fraction of a femtosecond.[8] SPPS and future XFEL's are accelerator-based sources that will usually provide only one of these pulses and therefore, are not self-synchronized. Since timing jitter between the pump and probe can degrade temporal resolution, synchronization of the x-ray source to an external ultrafast laser is a prime concern.

Laser-x-ray synchronization to within a few picoseconds can be achieved if the external laser repetition rate is tuned to a sub-multiple of the radio frequency (RF) which accelerates the electrons. To achieve higher resolution, we require a direct measurement of the relative time of arrival of the laser and the x rays, or the electrons that produced them, for every x-ray pulse. Although the precise time of arrival fluctuates from pulse to pulse, the recorded timing information can be used to reconstruct the ultrafast dynamics of the system.

Timing information in this experiment is obtained using a technique based on electro-optic sampling (EOS)[9] of the electric field surrounding the ultra-relativistic electrons that produce the x-ray pulses. Bunches of up to 2×10^{10} electrons are pre-compressed, accelerated through the 3km SLAC linear accelerator to an ultra-relativistic energy of $E = 28.5GeV$ ($\gamma = E/(mc^2) \approx 60{,}000$), and then compressed once more before delivery to the SPPS experiment. Electron trajectory simulations show that the ultra-relativistic bunch length at this point is as short as 12μm rms along the direction of motion, corresponding to 80 fs FWHM. As a consequence of Special Relativity, the Coulomb field of the electron bunch is nearly transverse to its motion. Its magnitude is the vector sum of the fields of the individual electrons. We estimate a peak electric field of hundreds of megavolts per meter at a few millimetres from the beam. This electric field induces a transient birefringence in a crystal, known as the electro-optic effect, which changes the polarization of laser light passing through the crystal at that instant. The electron bunch passes far from the crystal so that timing and pulse length can be measured *without* affecting the electrons.

To measure this effect, we sweep an ultrafast laser pulse across the EO crystal at an angle to the electron beam axis, as shown in figure 1. The top edge of the pulse intersects the crystal before the bottom edge, thus mapping time to the spatial coordinate across the diameter of the laser spot[10]. The angle of incidence of the laser probe

determines the sweep rate, and consequently the resolution of the measurement, while the laser beam diameter determines the duration of the measurement window. A polarizer transmits only the portion of the probe laser spot where the polarization was altered by the EO effect. The position of the signal indicates the time of arrival of the electron bunch, while its width and amplitude contain information on the temporal profile.

The time resolution of the measurement is limited by the thickness of the electro-optic crystal[11], the pulse length of the laser, and the spatial resolution of the imaging system used to monitor the transmitted laser pulse. For the measurements presented here we used a 200 μm thick <110> ZnTe crystal positioned 5 mm from the electron bunch trajectory. The electric field due to the bunch lies in the plane of the crystal. The 2 mm wide laser beam is incident at an angle of 45 degrees with respect to the crystal surface normal with an effective single-shot window of 9 ps.

To achieve precise timing, the EOS measurement and x-ray pump-probe experiments share a common laser system. A single Ti:sapphire oscillator is located in a laboratory next to the end of the x-ray beam line, approximately 150m from the EOS experiment. This large separation will be characteristic of future XFEL facilities. The laser oscillator operates at an actively-stabilized repetition rate of 102MHz, a sub-harmonic of the linear accelerator RF signal at SLAC and is phase-locked to this reference to better than 200 fs (rms integrated phase jitter from 1Hz to 40MHz); additional jitter and drift exists between the reference RF and the electron beam.

Ultrafast optical pulses from the oscillator are delivered to the EO crystal via a polarization-preserving single-mode optical fibre. Short optical pulses are required for EOS; however, ultrafast pulses broaden rapidly upon propagation in an optical fibre. For example, a 100 fs optical pulse will double its length after travelling only a few

centimetres due to spectral dispersion in the glass fibre. We overcome this by first spectrally dispersing the oscillator pulses using a series of gratings and a programmable spectral phase mask.[12] The required spectral phase shifts are programmed into the phase mask by a genetic learning algorithm.[13] The dispersion is opposite to that of the fibre, so that the pulses are recompressed as they travel through the fibre to the EOS experiment. Final compression occurs in glass optical elements just before the pulses encounter the EO crystal. In our experiments we delivered a 135 fs FWHM laser pulse to the EO crystal.

A series of EOS images of the electron bunch, taken as a function of bunch compression, are shown in Figure 2. These are the shortest single-shot electron bunch measurements to date.[14] The width of the EO signal reaches a minimum of 270 fs FWHM, which is a convolution of the instrument response and the electron bunch duration, and appears to be resolution limited. These data show the optimum compression settings and indicate that EOS could be used as a diagnostic in a future XFEL. A combination of a thinner crystal, a shorter laser pulse, and better imaging would be required to resolve the predicted 80 fs bunch duration at SPPS.

Twenty consecutive single-shot measurements made at SPPS are shown in figure 3. The centroid of each EO image can be determined with 30 fs accuracy, and indicates the time of arrival of the bunch with respect to the laser pulse. Random fluctuations from shot to shot are evident and were measured to be 194 fs rms over 100 s of observation.

There are additional sources of timing jitter between the EOS measurement and an SPPS x-ray-pump-probe experiment which are expected to be small. In order to determine the magnitude of this additional jitter we compared timing information measured using EOS with timing information from a laser-pump-x-ray probe study of

ultrafast non-thermal melting. The arrival time of the x-rays with respect to the pump laser could be determined to within 50 fs.[15] Data were collected over 30 s. The comparison is shown in figure 4. While each timing measurement method individually yields a jitter of approximately 200fs, the correlation shows only 60 fs rms jitter between the two techniques.

The short-term timing jitter at SPPS is approximately 200 fs. There are also long-term drifts, associated with the RF reference to which the laser oscillator is phase-locked and with the fibre laser transport system. Most pump-probe experiments, which require accumulation of data over multiple shots, would have their time resolution limited by these long-term drifts, which can be as great as 30 ps at SPPS. EOS could be used to identify and compensate for the sources of drift. Therefore, a continuous set of EOS measurements could be used to improve the time resolution of an SPPS x-ray pump-probe experiment from 30 ps to 60 fs or less.

Ultrafast research at XFEL's will require knowledge of the x-ray pulse arrival time with respect to a pump laser with femtosecond precision. This paper introduces the first technique that addresses this problem, and demonstrates sub-100 fs precision in the x-ray arrival time measurement. The time resolution for experiments at SPPS has been improved to 60fs rms, nearing the resolution limit imposed by the x-ray pulse duration. In principle, relative timing information from spatially resolved EO measurements could be extended to 1 fs, matching the projected performance of XFEL's into the foreseeable future.

---

1. Emma, P., Iverson, R., Krejcik, P., Raimondi, J., Safrarek, X. Femtosecond electron bunch lengths in the SLAC FFTB beamline. Contributed to IEEE Particle Accelerator Conference (PAC 2001), Chicago, Illinois, 18-22 Jun 2001, pp. 4038-4040.

2. Bentson, L., Emma, P., Krejcik, P. A new bunch compressor chicane for the SLAC LINAC to produce 30 fsec, 30KA, 20-GeV electron bunches. Contributed to 8th European Particle Accelerator Conference (EPAC 2002), Paris, France, 3-7 Jun 2002, pp. 683-685.

3. Wilke, I. *et al*. Single-shot electron-beam bunch length measurements. Phys. Rev. Lett. **88,** 124801 (2002).

4. Spielmann, C. *et al*. Generation of coherent x-rays in the water window using 5 fs laser pulses. *Science* **278**, 661-664 (1997).

5. Chang, Z. H., Rundquist, A., Wang, H. W., Murnane, M. M., & Kapteyn, H. C. Generation of coherent soft X rays at 2.7 nm using high harmonics. *Physical Review Letters* **79,** 2967-2970 (1997).

6. Rischel, C. *et al*. Femtosecond time-resolved x-ray diffraction from laser-heated organic films. *Nature* **390,** 490-492 (1997).

7. Schoenlein, R. W. *et al.* Generation of femtosecond pulses of synchrotron radiation. *Science* **287**, 2237 (2000).

8. Hentschel, M. *et al*. Attosecond metrology. *Nature*. **414**, 509-513 (2001).

9. Valdmanis, J. A. & Mourou, G. Sub-picosecond electro-optic sampling – principles and applications. *IEEE J. Quantum Electron.* **22,** 69-78 (1986)

10. Shan, J. *et al*. Single-shot measurement of terahertz electromagnetic pulses by use of electro-optic sampling. *Optics Lett.* **25,** 426 (2000).

11. Wu, Q., Zhang, X. C. Ultrafast electro-optic field sensors. *App. Phys. Lett.* **68,** 1604-1606 (1996).

12. Lee, S. L., Cavalieri, A. L., Fritz, D. M., & Reis, D. A. Adaptive dispersion compensation for remote fiber delivery of NIR femtosecond pulses. *Optics Lett.* accepted for publication.

13. Baumert, T., Brixner, T., Seyfried, V., Strehle, M., & Gerber, G. Femtosecond pulse shaping by an evolutionary algorithm with feedback. *App. Phys. B.* **65,** 779-782 (1997).

14. Berden, G. *et al.* Electro-optic technique with improved time resolution for real-time nondestructive, single-shot measurements of femtosecond electron bunch profiles. *Phys. Rev. Lett.* accepted for publication

15. Sokolowski-Tinten, K. *et al*. First ultrafast x-ray measurements at SPPS. Contributed to International Quantum Electronics Conference (IQEC 2004) San Francisco, CA, 16-21 May 2004. IPDB3.

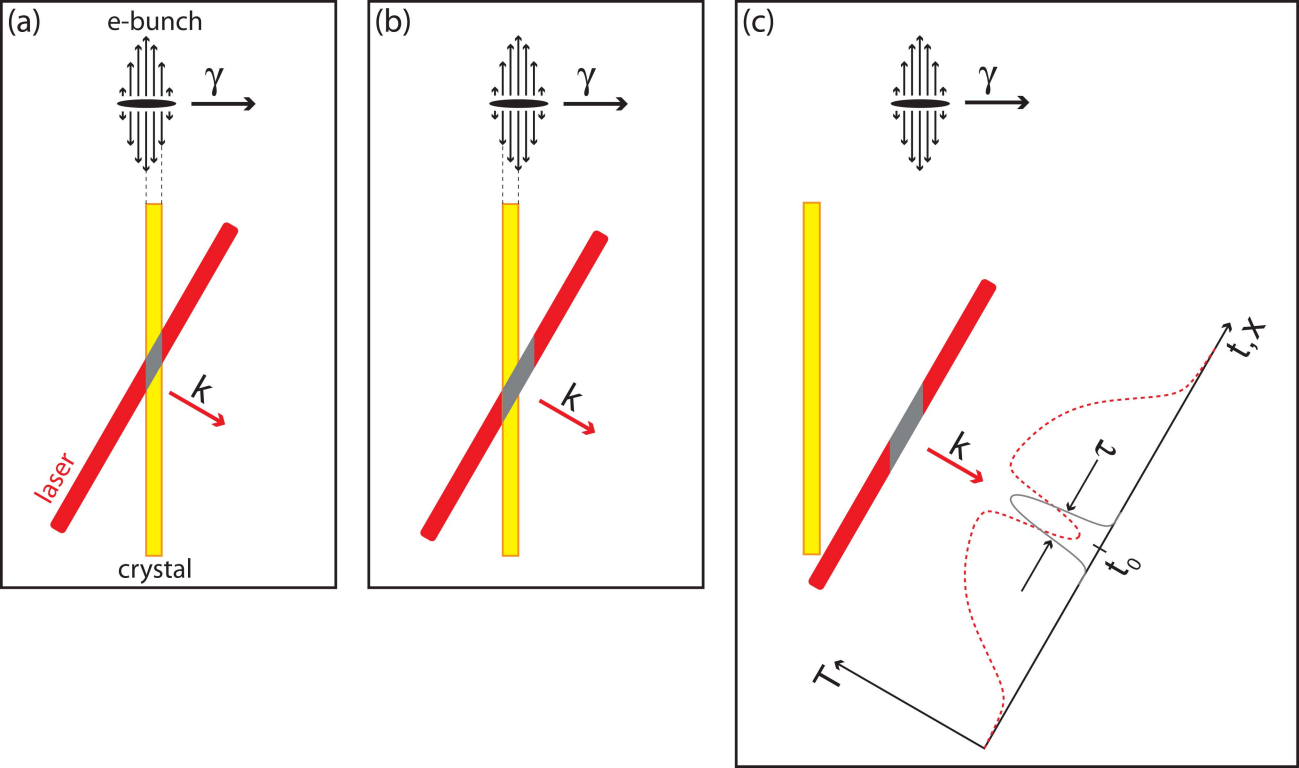
Acknowledgments: Portions of this research were carried out at the Stanford Synchrotron Radiation Laboratory, a national user facility operated by Stanford University on behalf of the U.S. Department of Energy, Office of Basic Energy Science and the USDOE at UC LLNL contract W-7405-Eng-48.

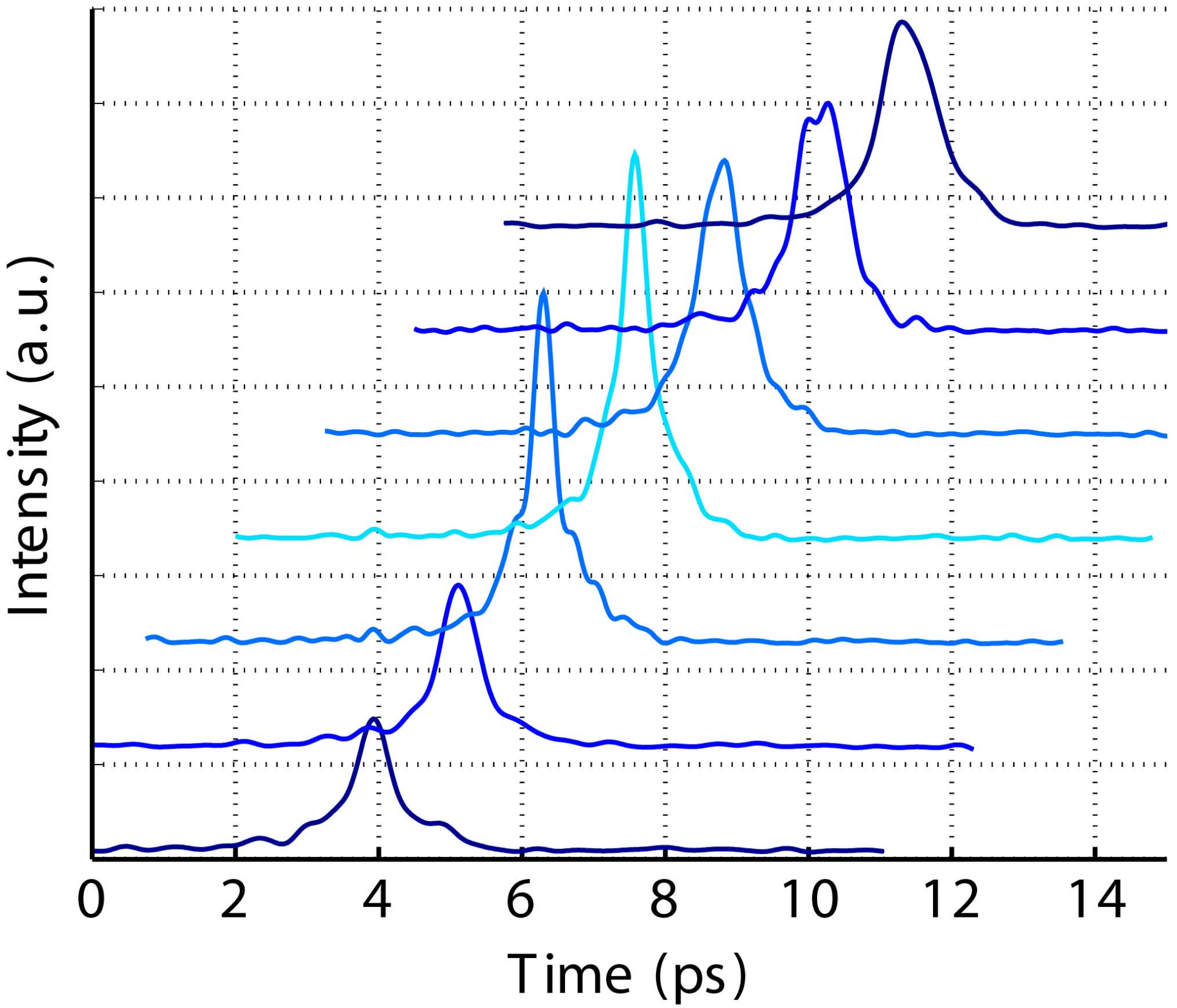
Figure 1: Cartoon depiction of spatially resolved electro-optic sampling. The frames represent three instants during the course of the measurement. The yellow object represents the EO crystal. The red object represents an ultrafast laser probe pulse moving from top-left to bottom-right. The black oval represents the ultra-relativistic electron bunch moving from left to right (with electric field lines indicated). In (a) the front of the electron bunch and the laser pulse interact in the EO crystal. In (b) the back of the electron bunch interacts with the laser pulse. In (c) the electron bunch has passed the crystal and its shape has been imprinted on the laser polarization profile, mapping time into space. The intensity profiles of the laser polarization components are plotted. The width of the signal, τ, represents a convolution of the electron bunch length, the crystal EO response, and the laser pulse duration. The centroid of the signal indicates the relative time of arrival.

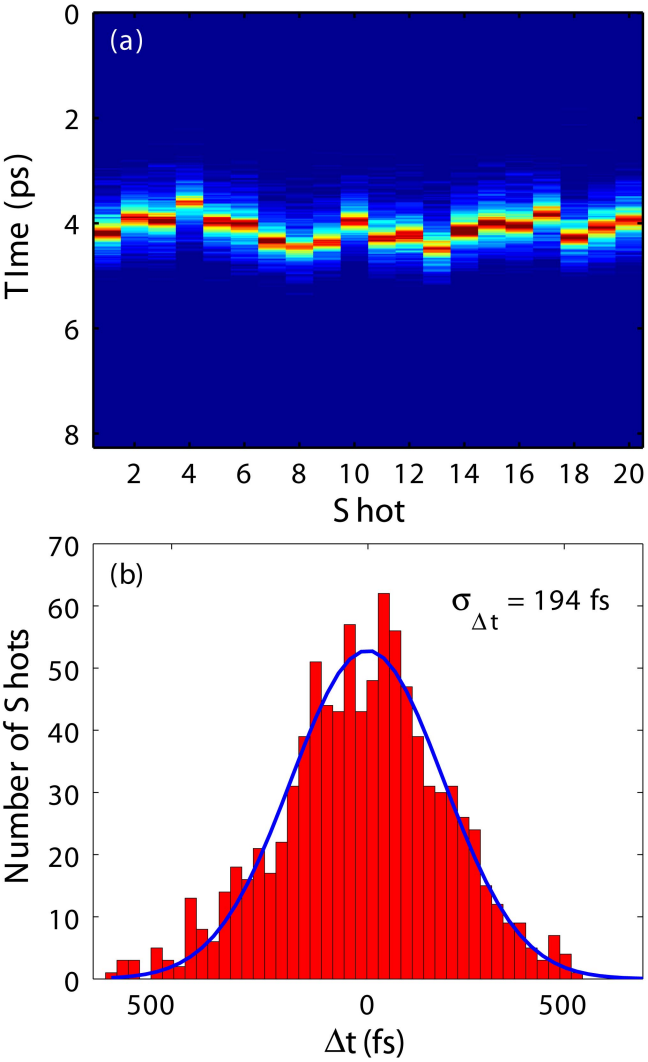
Figure 2: A series of electro-optic signals (offset for display) collected as the electron bunch compressors at SLAC were changed, changing the bunch length. The minimum FWHM shown represents a time of ~270 fs which is the resolution limit of the apparatus.

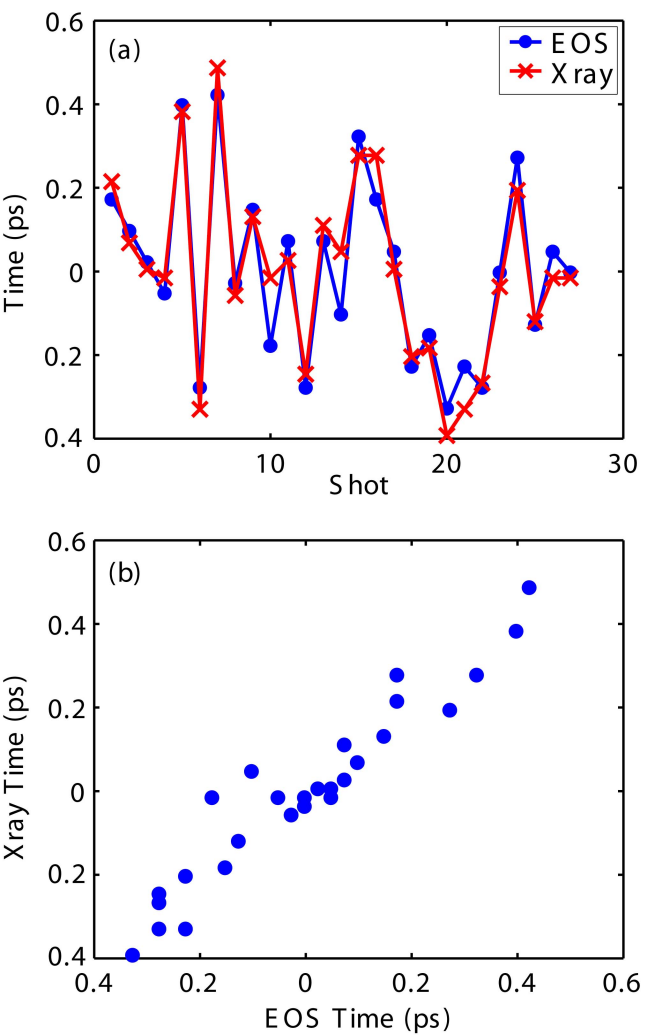
Figure 3: (a) Twenty consecutive single shot electron bunch measurements. The bright band in each column is the electro-optic signal, its location indicates the time of arrival of the electron bunch with respect to the laser probe pulse, and its width corresponds to the electron bunch duration. (b) Normalized arrival time histogram of 1000 consecutive single shots.

Figure 4: (a) Shot by shot comparison of pulse arrival times determined from EOS measurements (blue), and from ultrafast laser-pump-x-ray-probe measurements (red). Both measurement techniques show a shot-to-shot jitter of ~200 fs. (b) Correlation plot of the data shown in (a). The relative jitter between the two techniques is ~60 fs.


(a)
e-bunch
γ
k
laser
crystal
(b)
γ
k
(c)
γ
k
τ
t_0
t,x
T


Intensity (a.u.)
0
2
4
6
8
10
12
14
Time (ps)


(a)
TIme (ps)
0
2
4
6
8
2
4
6
8
10
12
14
16
18
20
S hot
(b)
σ Δt = 194 fs
Number of S hots
70
60
50
40
30
20
10
0
500
0
500
Δt (fs)


(a)
EOS
X ray
Time (ps)
Shot
(b)
Xray Time (ps)
EOS Time (ps)